Self-Sustaining Breeding in Advanced Reactors: Characterization of Natural Resources

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Glossary

Ac Actinides, elements with atomic numbers 89 to 103.

Actinides Eigen-composition Stabilized actinides composition, an equilibrium composition which results from a long irradiation of one parent nuclide or their mixture. Necessary condition for the stabilization is that the mass of parent nuclide/s and the irradiation flux do not vary strongly. It is specific and inherent feature for every reactor and every parent nuclide/s and it represents the Eigen-vector of the respective Bateman equation.

B&B Breed-and-Burn; a mode of open cycle operation, where the fissile excess bred during irradiation is equal or higher than the discharged fissile mass after the irradiation. The reactor burns its own bred fuel. The discharged fuel does not need to be recycled and the fresh fuel does not need to contain primordial or synthetic fissile nuclides.

Binding energy per nucleon Energy normalized per nucleon, which would be released when nuclei would be synthetized from protons and neutrons or which would be needed to disassemble the nucleus into protons and neutrons.

Breeding Transmutation of actinide nuclide initiated by neutron capture, which increases fission probability. Typically, primordial non-fissile (fertile) nuclides ^{232}Th and ^{238}U are transmuted into synthetic (man-made) fissile ^{233}U and ^{239}Pu.

Burnup Share of already fissioned actinides. It can be expressed in FIMA % (Fissions per Initial Metal Atom) or as energy released from given mass of actinides in MWd/kg (Megawatt days per kilogram). Since the energy per fission is approximately the same for all actinide nuclides, values expressed in these two units are proportional and roughly 10 times higher for the second unit.

Closed cycle A chain of process steps which is closed for the respective working medium. The medium, in nuclear fuel cycle the actinides, is recycled and does not extensively leave the chain.

Cross-section A measure of the probability that neutron will interact when flying by with the respective nucleus (microscopic cross-section) or when fling through with the respective nuclei concentration (macroscopic cross-section).

Decay series Series of consecutive radioactive decays and intermediate decay products, which are needed to reach the final stable nuclide. All actinides are radioactive and decay through one of the four existing actinides alpha decay series.

Fertile nuclide Nuclide which can be transmuted into fissile nuclide.

Fission barrier It represents the minimal added energy by the interacting particle to cause nuclear fission.

FPs Fission Products, typically two fragments of actinides fission.

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Irradiation chain Series of consecutive transmutations and of the respective daughter products induced by neutron irradiation of one parent nuclide, e.g., ^{232}Th or ^{238}U. The radioactive decays of the daughter products are part of the chain.

MCFR Molten Chloride Fast Reactor, unmoderated MSR type based on molten chlorides.

MSR Molten Salt Reactor, a reactor in which the molten salt has substantial function, in this chapter the label is used for graphite moderated fluoride salt based MSR.

Neutron economy Balance between neutron generation and neutron losses in a reactor. Good neutron economy has high neutron generation and small neutron losses by parasitic absorption and leakage from the reactor.

Open cycle A chain of process steps which is open for the respective working medium. The medium, in nuclear fuel cycle the actinides, is not recycled and leaves extensively the chain.

Primordial actinides Actinide nuclides, presumably originated by rapid neutron capture process during a supernova explosion, which have long enough half-life for radioactive decay to be still present on the earth.

Self-sustaining breeding Breeding process where breeding rate $\geq$ fission rate, the mass of fissile synthetic nuclides is conserved in the reactor, and fission chain reaction is sustained solely by these synthetic nuclides. A long time-horizon is being considered in this article.

Synthetic actinides Actinide nuclides originated, typically by neutron-induced transmutation, from primordial actinides. Synthetic nuclides are, by definition, a much broader group than minor actinides or trans-uranic elements. For thorium and uranium elements, both primordial and synthetic isotopes exist.

Waste Waste is an unwanted side product, which is further unusable. In the nuclear fuel cycle, synthetic actinides represent a side product, which is reusable. However, they can be declared as a waste by nuclear fuel cycle policy or when it is too laborious to recover them, e.g., from reprocessing losses of certain fuel forms.

Introduction

This is the first of a series of three chapters that is dedicated to the identification of the advanced reactor technologies that are capable of self-sustaining breeding when fueled with either ^{232}Th or ^{238}U, and to the investigation of equilibrium fuel cycles properties and neutronics performance. This first chapter characterizes the available fuel resources from the nuclear and reactor physics perspective. It presents in detail the features of the ^{232}Th and ^{238}U irradiation chains using two advanced reactors adopted from (Losa, 2016; Krepel and Losa, 2019) as an example and the knowledge of equilibrium nuclides concentration and reaction rates is used for characterization of neutron economy and ingestion radiotoxicity related to these chains. The second chapter (Krepel and Losa, 2021) determines the equilibrium fuel composition in advanced reactors and compares the basic features of the equilibrium fuel cycle and selected reactor neutronic performance characteristics at the equilibrium state. The third chapter (Krepel, 2021) evaluates the fuel cycle performance from a neutronics perspective by means of the earlier obtained equilibrium parameters. It provides an additional insight on why some advanced reactor concepts can act as self-sustaining breeders in closed or even open fuel cycle, while others do not.

Actinides as a resource

In terms of producing energy by atomic nuclei transmutation, the nuclear fission chain reaction is the only alternative to nuclear fusion. From the resources available on earth only actinides can be utilized in nuclear fission reactors. They were presumably originated by rapid neutron capture processes in the core of a collapsing star and expelled into space by gravitational shock waves during its supernova explosion. Hence, the nuclear fuel cycle relies on a resource, which is not renewable. Three dominating primordial actinide nuclides are present on earth: ^{235}U, ^{238}U and ^{232}Th. Their estimated reserves somewhat follow their half-lives for radioactive decay (see **Fig. 1**). Accordingly, there is probably three to four times more thorium than uranium, and the ^{235}U share in natural uranium is limited to 0.7%. The ^{235}U reserves are the smallest; at the same time, it is the only primordial nuclide fissile by thermal neutrons. Therefore, it can be directly utilized in nuclear fission reactors. The other two primordial nuclides can also be utilized, albeit not directly and not in all reactors. Accordingly, they are foreseen as a resource for advanced reactors (Stanculescu, 2021).

Primordial and synthetic actinides

Actinides nuclides can be divided into primordial and synthetic. The three major primordial nuclides have half-lives of the same order of magnitude as the age of earth (see **Fig. 1**). The synthetic actinides are originated by primordial nuclides transmutation, typically by neutron capture reactions. Fission reactions also represents nuclear transmutation; however, the resulting Fission Products (FPs) do not belong to the chemical group of actinides.

Fission and transmutation

The ^{235}U reserves are the smallest; at the same time, ^{235}U is the only primordial nuclide suitable for establishing a fission chain reaction. Consequently, vast majority of current nuclear power plants rely on enriched uranium and fission predominantly ^{235}U.

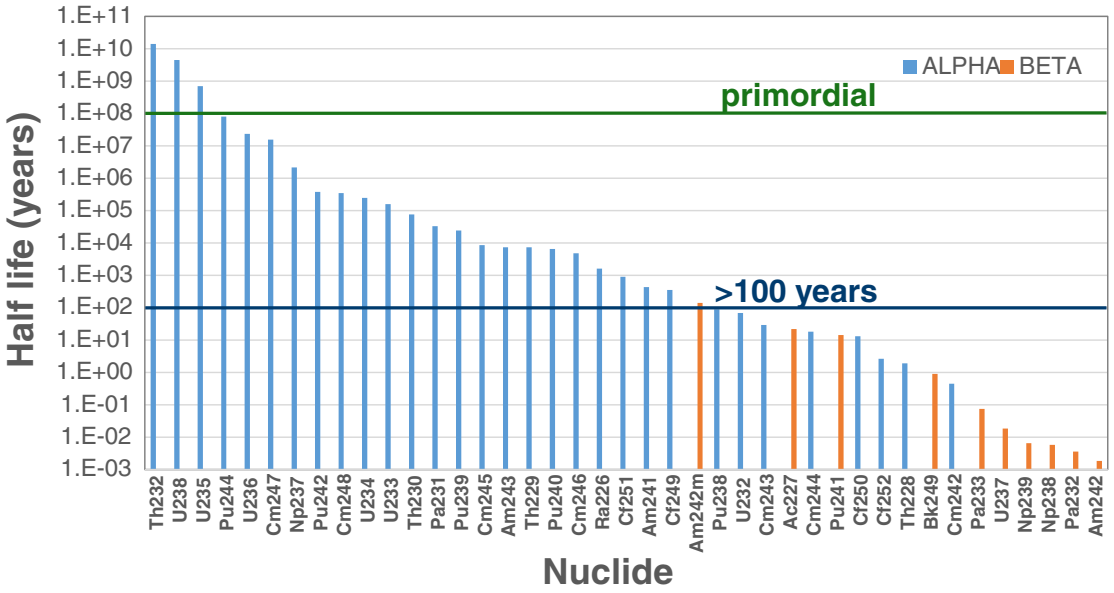

Fig. 1 Half-life and type (alpha or beta) of radioactive decay for primordial and synthetic actinides.

The consumption of natural uranium in these reactors does not exceed 1%. If also the thorium reserves are considered, not more than 0.2% of primordial actinides are utilized for energy production in ^{235}U-fueled reactors. Moreover, the discharged fuel from these reactors contains synthetic actinide nuclides, e.g., ^{234}U, ^{236}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Am, originated from ^{235}U and ^{238}U irradiation. Synthetic actinides are the major cause of the long-term stewardship burden related to the final spent fuel repository. Their recycling in ^{235}U-fueled reactors is possible, but only to a limited extent. Since the fuel is usually not recycled, these reactors are operated in so-called open cycle (Crawford, 2021).

Already in 1940 it was discovered that transmutation of fertile ^{238}U results in fissile ^{239}Pu (Seaborg, 1994). Equally, fertile ^{232}Th can be transmuted into fissile ^{233}U. This transmutation process is called breeding and reactors relying on this process are called breeders. Once originated, the synthetic nuclides ^{239}Pu and ^{233}U can sustain the fission chain reaction and the breeding process itself. Accordingly, they practically act as a catalyzer for energy production from the primordial nuclides ^{238}U and ^{232}Th. Even though the first ^{239}Pu was generated by a different (particle accelerator based) nuclear process, the fission chain reaction driven by ^{235}U is the most natural option to generate the initial synthetic nuclides and to launch the self-sustaining breeding. However, not all advanced reactors can sustain this process, and some perform better than others. The aim of this article is to provide insight from a nuclear and reactor physics perspective, as to why it is so.

Natural resources utilization

The amount of primordial actinides on earth is finite and not renewable. Accordingly, the sustainability of the nuclear fuel cycle is determined by its capability to maximize its natural resources utilization. As already mentioned, utilization of natural uranium does not exceed 1% in ^{235}U-based open cycle and it can reach up to 20–35% in the breed-and-burn (B&B) open cycle mode (Qvist, 2021; Krepel and Kramer, 2021; Greenspan, 2021). Nonetheless, the highest resources utilization can be achieved by breeding in a closed cycle (Wigeland, 2021; Krepel et al., 2018). The cycle is called closed, because actinides as the working medium are recycled and do not significantly leave the cycle. Achievable utilization in this fuel cycle is limited solely by the recycling losses (losses in fuel reprocessing and refabrication) and fuel burnup per cycle (see **Fig. 2**). It can reach up to 95–98% of the resources. Discharged fuel burnup influences the losses, because fuel with higher burnup is less often recycled. In an ideal reactor, the discharged fuel burnup should be 100% FIMA and reprocessing frequency zero. Should this be possible, breeding in closed cycle and in the B&B open cycle will become identical. Even though unrealistic, it may be approached in reactors with liquid fuel from which solely FPs are extracted from the core.

The reason why not all reactor concepts are capable of being self-sustaining (isobreeding/break-even) breeders is the tight neutron economy. Simple fuel balance of a breeder reactor requires that for each fissioned synthetic nucleus at least one transmutation of primordial nuclide takes place. Accordingly, up to two neutrons are needed from each fission: one for sustaining the chain reaction and one for maintaining of the fissile material balance. The neutron economy of a breeder is thus very tight. Less than one neutron per fission should be lost due to the neutron leakage or parasitic neutron capture in the coolant and structural materials or in the fuel itself. Furthermore, once discharged from a breeder, the irradiated fuel must be recycled to maintain the fissile material balance. The only exception from this requirement are very strong breeders, where the fissile excess bred during the irradiation would be equal or higher than the discharged fissile mass after the irradiation. Since the bred excess is not necessary for the fissile

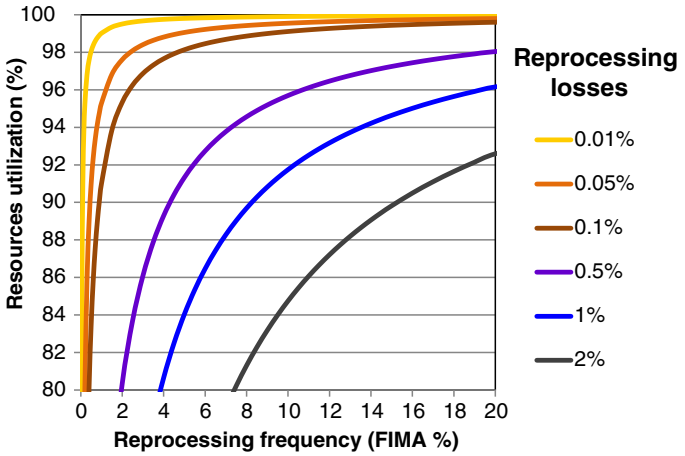

Fig. 2 Natural resources utilization as a function of reprocessing frequency (inversely proportional to fuel burnup per cycle) and losses in recycling. From Krepel J, Hombourger B and Losa E (2018) Fuel cycle sustainability of Molten Salt Reactor concepts in comparison with other selected reactors. *PHYTRA4, Marrakech, Morocco, September 17-19.*

mass conservation, these very strong breeders can self-sustain the breeding in open cycle in the B&B mode (Qvist, 2021). The name is derived from the fact that the loaded fuel does not need to contain fissile actinides; they are bred during the irradiation in the B&B reactor and suitable fraction of them fission in that reactor.

Catalyzer and waste, simultaneously

Synthetic actinide nuclides are the necessary catalyzer for the primordial actinides utilization. However, in the open fuel cycle practiced today, or as losses of recycling in a closed cycle, they cause a long-term stewardship burden. Their half-lives are too short to be primordial, but too long to disappear swiftly once originated (see **Fig. 1**). Furthermore, synthetic actinides decay in chains and one fast decaying nuclide can be a source of another slow decaying nuclide and vice-versa. The social and environmental sustainability considerations require minimal long-term stewardship burden; hence, the amount of synthetic actinides in the waste stream should be minimized. The burden should be measured in terms of their long-term radiotoxicity per unit of produced energy. Indefinite recycling in closed cycle provides the ultimate solution not only for maximizing resources utilization, but also for minimizing waste production. The recycling losses define the achievable limit for both measures. In the open cycle, all discharged actinides are considered as a waste and the policy of many countries is to seal them forever in a spent fuel repository without recycling. However, synthetic actinides are in reality a resource and they are treated as such in this article.

Irradiation chains of primordial actinides

Since any synthetic actinide, as a transmutation product, can undergo another transmutation, the irradiation of primordial actinides by neutrons results in a chain of synthetic actinides. The irradiation chains of ^{232}Th and ^{238}U are especially relevant for natural resources utilization.

Irradiation chains

For each reactor and for each primordial nuclide, the composition of generated synthetic actinides tends to stabilize and reaches equilibrium concentration after sufficiently long neutron irradiation. Necessary condition for the composition to converge to its equilibrium state is that the mass of irradiated nuclide and the neutron flux do not vary strongly. Hence, when the same fuel is repetitively irradiated in closed cycle, soon or later the equilibrium composition of synthetic actinides will be established. This stabilized actinides composition is actually the Eigen-composition of the respective Bateman equation (Bateman, 1910) and it is specific and inherent feature of given reactor and parent primordial nuclide. For the illustrative purposes, the ^{232}Th and ^{238}U irradiation chains in thermal and fast neutron spectra are presented in **Figs. 3–6**. The respective reactors used for the simulation are graphite moderated Molten Salt Reactor (MSR) with fuel in form of eutectic mixture of lithium fluoride and actinides tetra-fluoride salts and Molten Chloride Fast Reactor (MCFR) with fuel in form of eutectic mixture of sodium chloride and actinides tetra-chloride salts. For more detail about the selected reactors and modeling assumptions refer to (Krepel and Losa, 2021).

Even in an open cycle, the fuel composition tends to converge to equilibrium. The ^{238}U mass does not vary strongly in the ^{235}U-fueled reactors. Already at the end of an open-cycle irradiation, the immediate daughter product ^{239}Pu is practically at equilibrium concentration and its daughter ^{240}Pu close to it. The concentrations of higher plutonium isotopes are not yet converged, and

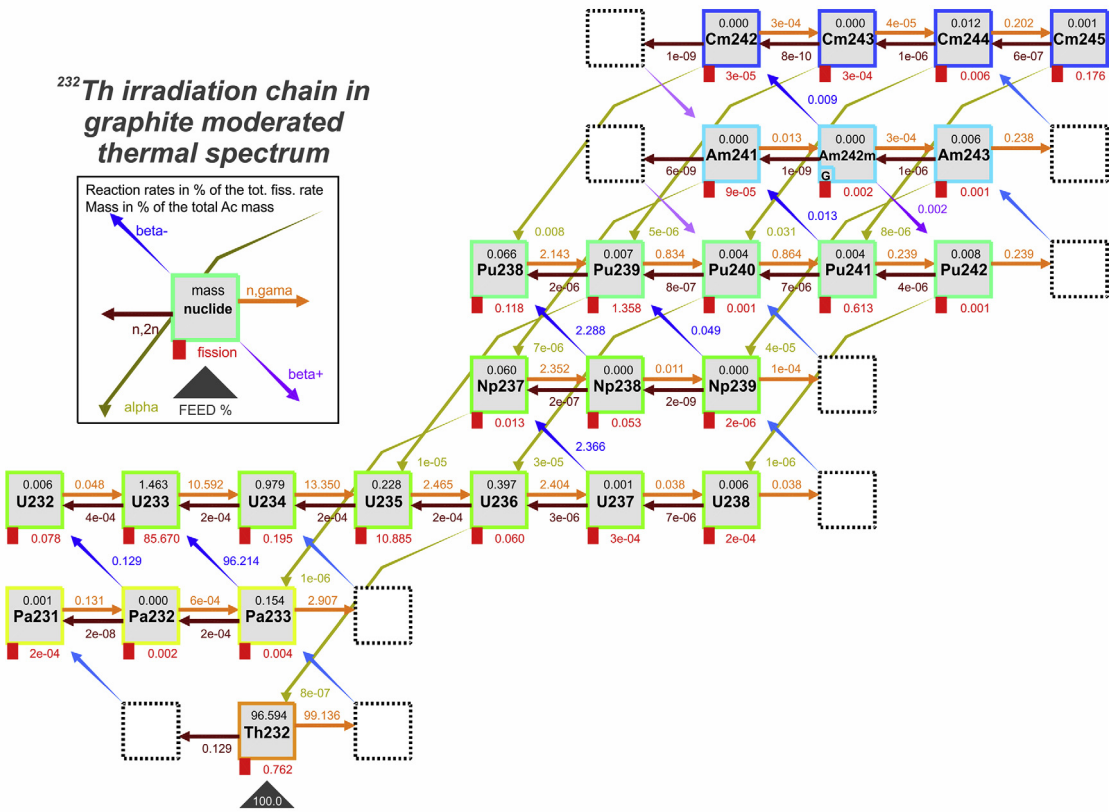

Fig. 3 Example of the ^{232}Th irradiation chain with relative masses and reaction rates in graphite moderated MSR. It partly illustrates also the ^{235}U irradiation chain. From Losa E (2016) *U-Pu and Th-U Fuel Cycle Closure*, PhD thesis. Prague: Czech Technical University in Prague, Department of Nuclear Reactors.

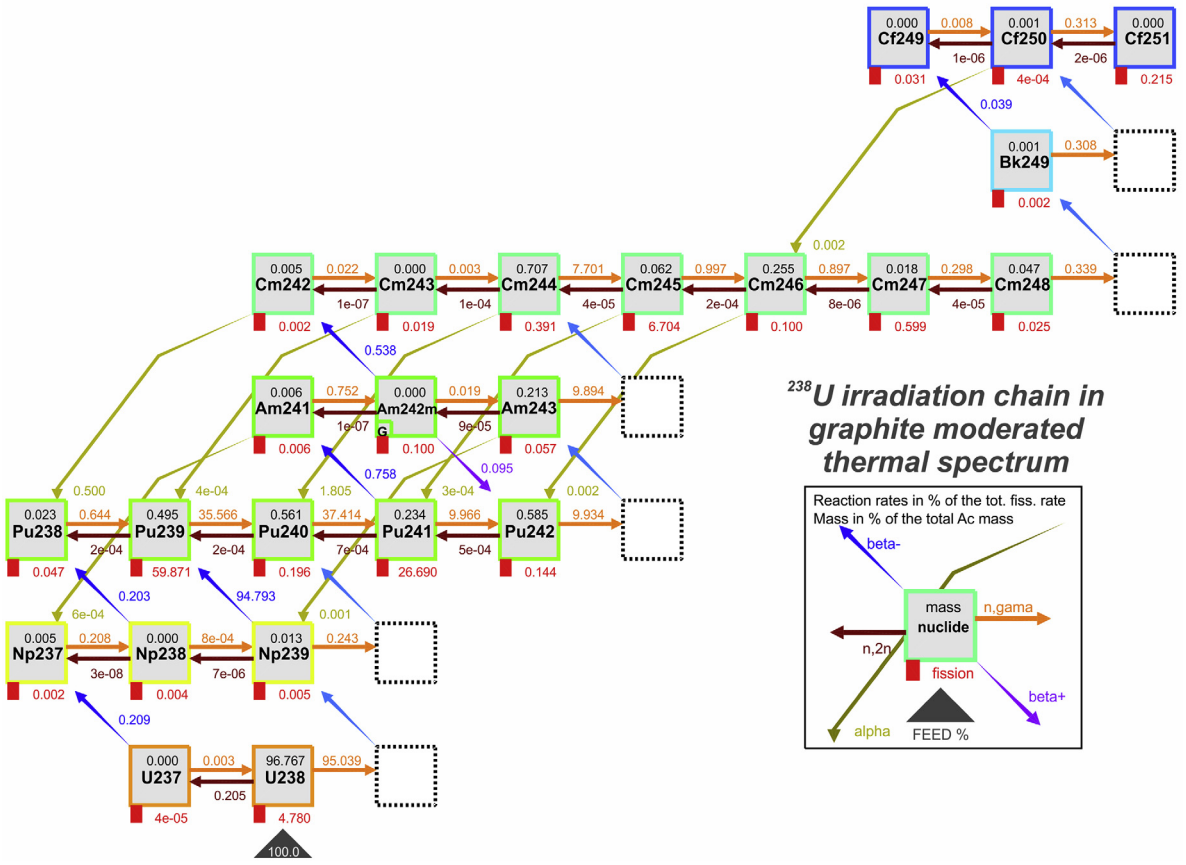

Fig. 4 Example of the ^{238}U irradiation chain with relative masses and reaction rates in graphite moderated MSR. From Losa E (2016) *U-Pu and Th-U Fuel Cycle Closure*, PhD thesis. Prague: Czech Technical University in Prague, Department of Nuclear Reactors.

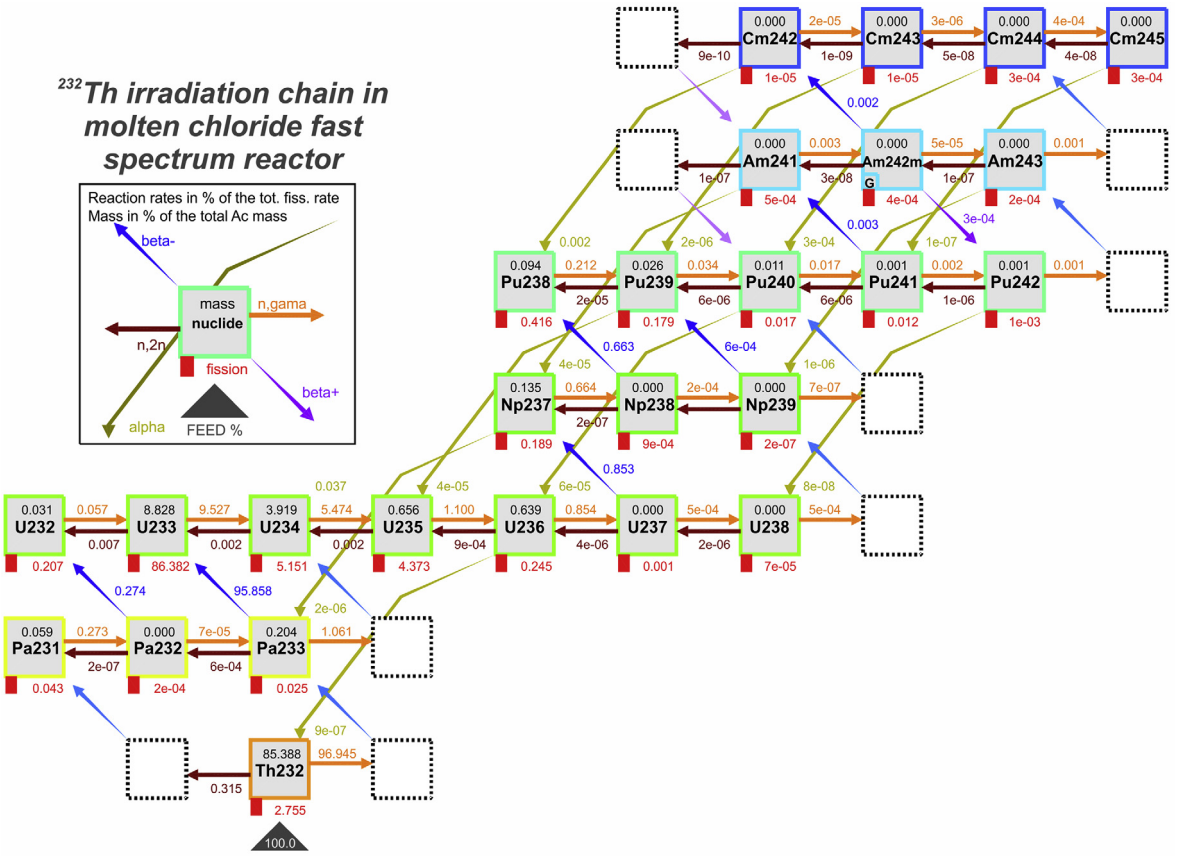

Fig. 5 Example of the ^{232}Th irradiation chain with relative masses and reaction rates in MCFR. It partly illustrates also the ^{235}U irradiation chain. From Losa E (2016) *U-Pu and Th-U Fuel Cycle Closure*, PhD thesis. Prague: Czech Technical University in Prague, Department of Nuclear Reactors.

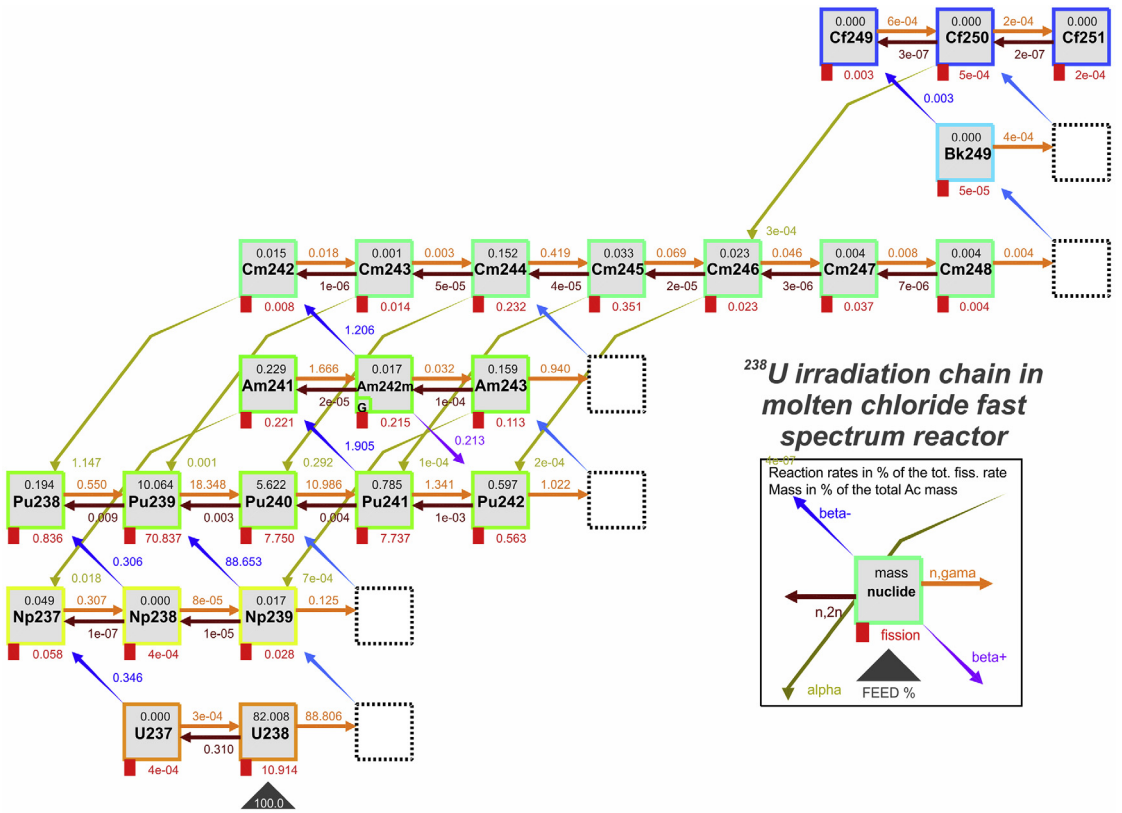

Fig. 6 Example of the ^{238}U irradiation chain with relative masses and reaction rates in MCFR. From Losa E (2016) *U-Pu and Th-U Fuel Cycle Closure*, PhD thesis. Prague: Czech Technical University in Prague, Department of Nuclear Reactors.

americium, curium, and their daughters will need substantially longer time for stabilization. The convergence process of higher plutonium isotopes is also the reason why plutonium is recycled optionally only once in ^{235}U-fueled reactors (although a second and third plutonium recycling is being considered in France (Guillaume et al., 2018)). The plutonium multi-recycling in ^{235}U-fueled reactors will not substantially increase the resources utilization; at the same time, it surely helps to minimize the synthetic actinides amount in the waste stream.

During the irradiation of ^{235}U, concentrations of daughter products like ^{234}U, ^{236}U and ^{237}Np also tends to equalize quite fast. Nonetheless, ^{235}U is fissile and its own concentration strongly decreases during the irradiation. Hence, equilibrium as such is never reached. Even though it is not accurate, the basic features of the ^{235}U irradiation chain can be understood from the ^{232}Th chain, because it is its sub-set. Based on the example in **Fig. 3**, for instance, the equilibrium concentration of ^{236}U seems to be higher than the ^{235}U concertation in a thermal spectrum reactor. However, also in fast neutron spectrum (see **Fig. 5**) the ^{236}U concentration is similar to ^{235}U concentration. The irradiated uranium, with left-over ^{235}U and its daughter products ^{234}U and ^{236}U can be also recycled in ^{235}U-fueled reactors. In this case it needs to pass again through the enrichment process to reduce the ^{238}U amount.

Nuclear physics implications

There is some obvious symmetry in the irradiation chains presented in **Figs. 3 and 4** or in **Figs. 5 and 6**. This symmetry, but also all other features of nuclides such as: type of radioactive decay, half-lives, absorption cross-section, fission-to-capture probability, or even the energy released per fission can be explained by nuclear physic and, in particular, by the respective binding energy per nucleon and by the fission barrier.

Binding energy and line of stability

The binding energy per nucleon (protons and neutrons in the nucleus) is well described by empirical liquid drop model or actually the Weizsäcker formula (Weizsäcker, 1935). This formula has five terms: volume, surface, Coulomb, asymmetry and pairing. Balance of these terms define a line of stability. At this line or close to it the binding energy per nucleon is the highest and the nuclei are most stable. The line is defined by an interplay of only three terms of the Weizsäcker formula, because the volume term has no impact on the stability and the pairing term causes only local perturbations. The surface term is important for light nuclei and plays a role in fusion reactions but is less relevant for heavy nuclei and their fission. Furthermore, for nuclei, with the same number of nucleons, which are located on a diagonal (roughly orthogonal to the stability line in **Fig. 7**), the surface term is the same. The highest binding energy on this diagonal is determined by the balance of the Coulomb and asymmetry terms. These two terms quantify the reduction of binding energy caused by proton repulsive forces and by asymmetry between total proton and neutron count, see also Napolitano (2021) and Klay (2021).

Repetitiveness of nuclei properties

The Coulomb repulsive forces in actinide nuclei, with more than 89 protons, are enormous. The higher neutron count reduces the proton repulsive force. At the same time, it weakens the binding energy through the asymmetry term. Maximal stability of actinide nuclei is reached at 1.55–1.6 neutrons per proton. Yet they are all unstable and undergo radioactive decay. The approximate line of stability shown in **Fig. 7** follows a trend where the same stability is obtained when addition of one proton is accompanied by addition of two neutrons. Nonetheless, with increasing proton number, the stability of actinides generally decreases. Moreover, due to the pairing effect, nuclei with even number of neutrons and/or protons have higher binding energy. Accordingly, properties of the nuclei along the stability line are repeating with steps of 2 protons and 4 neutrons (+2p, +4n). This repetition, illustrated in **Table 1**, is the major reason why exactly ^{232}Th and ^{238}U are present on earth and why there is a symmetry of their irradiation chains. Based on this repetition, ^{244}Pu could have been the third primordial actinide nuclide. Unfortunately, its half-life for radioactive decay is 56 times shorter than for ^{238}U. Nonetheless, for illustration of the repetitiveness, it is included in **Table 1**.

Nuclides in the ^{232}Th irradiation chain have less protons and are generally more stable. The respective longer half-lives are the reason why the estimated ^{232}Th reserves are higher and why the initial radiotoxicity of the respective synthetic actinides seems lower (refer to the later discussion). Nonetheless, the longer half-lives also result in a 11 times higher ^{233}Pa concentration than the respective ^{239}Np concentration in the ^{238}U chain and increase so the parasitic neutron capture probability. The second major difference between the ^{232}Th and ^{238}U irradiation chains is the very short half-life of ^{241}Pu and its different type of decay. At the corresponding position in the ^{232}Th chain, there is α decaying ^{235}U with the third longest half-life. As illustrated in **Fig. 7**, the major irradiation chain can branch at the ^{241}Pu position. Dominant direction after this split depends on the ratio of decay and capture rates. In a thermal neutron spectrum, in which the ^{241}Pu capture cross-section is larger and its resulting concentration is thus smaller, the capture rate dominates over the decay rate (see **Figs. 3 and 4**). In a fast neutron spectrum it can be the opposite (see **Figs. 5 and 6**).

Fission probability and cross-section amplitude

The fission barrier, as the second nuclear physics parameter, is not presented here graphically, but in the area of interest it has slight slope (Petermann et al., 2012); it decreases with increasing number of protons and decreasing number of neutron in the nucleus.

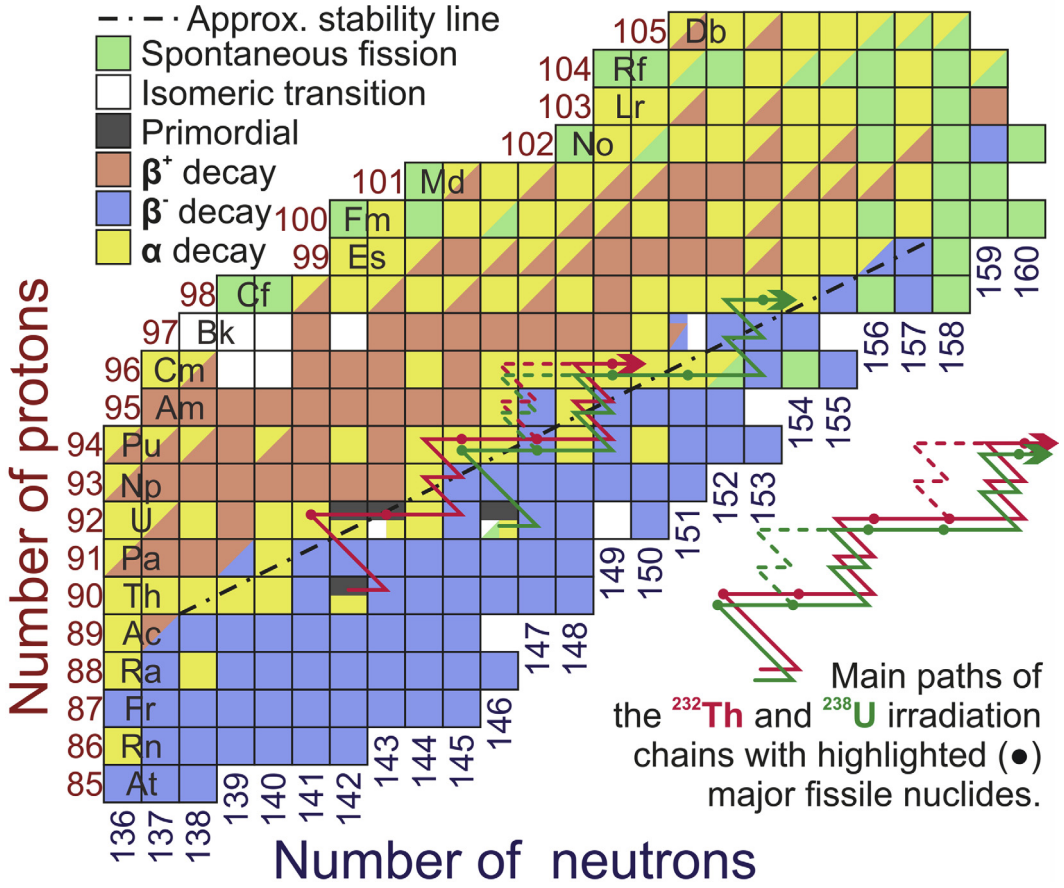

Fig. 7 Local nuclide chart of the actinides area with highlighted approximated line of stability and major paths for the ^{232}Th and ^{238}U irradiation chains with highlighted major fissile nuclides.

The Coulombian repulsive forces get stronger and the fission barrier weaker, when more protons are closer to each other (less neutrons). This is partly the reason why ^{233}U (see Fig. 7) has higher fission-to-capture probability than ^{235}U or ^{239}Pu. At the same time, ^{233}U and ^{235}U as the major fissile nuclides in ^{232}Th chain have lower number of nucleons than the respective ^{239}Pu in ^{238}U chain and release so, in average, less neutrons per fission.

To fission, each nucleus needs to overcome the fission barrier. The required energy is provided by the interacting neutron in potential (its binding energy) and kinetic form (its collision speed). Practically all actinide nuclides can be fissioned by fast neutrons with a certain probability. From this perspective, ^{238}U can be fissioned at lower neutron speed than ^{232}Th (see the respective fission rates in Table 3), what represents certain advantage for the ^{238}U irradiation chain. Possibility of fission by thermal neutrons is determined solely by the balance between fission barrier and binding energy. Since the binding energy depends also on the pairing effect, nuclei with odd number of neutrons are usually fissile by thermal neutrons.

The pairing effect is also relevant for the absolute size of neutron absorption cross-section. Nuclei with even number of neutrons have typically lower cross-sections for interaction with neutrons. This is actually the reason why the concentration of fissile nuclide in self-sustaining breeders is one or two orders of magnitude lower than the concentration of fertile nuclide.

Neutron economy

In the equilibrium irradiation chain, see Figs. 3–6, the actinides concentrations and reaction rates are stabilized. As a result, the equilibrium chain provides many perspectives and options allowing for estimating its neutron economy. The basic question is whether the reactor core could be critical with such an equilibrium fuel composition. Since all reaction rates in the presented chains are normalized to the fission rate and shown in percent, they can be interpreted as the history of one neutron generation, which was born form 100 fissions.

Table 1 Illustration of actinides properties repetitiveness along the line of stability with the steps of 2 protons and 4 neutrons for ^{232}Th, ^{238}U and ^{244}Pu irradiation chains.

Nuclide chains											
1	*2*	*3*	*Position*	*Protons (p)*	*Neutrons (n)*	*Fissile*[a]	*Decay*	*Longest half-life*	*Half-life ratio 1:2*	*Half-life ratio 2:3*	*Comment*
^{232}Th	^{238}U	^{244}Pu	Starting points	even	even	not	α	1.4E+10 years	3.1	56	b
^{233}Th	^{239}U	^{245}Pu	+0p,+1n	even	odd	–	β^-	<day	1	0.03	
^{233}Pa	^{239}Np	^{245}Am	+1p,+0n	odd	even	not	β^-	27 days	11	28	
^{233}U	^{239}Pu	^{245}Cm	+2p,-1n	even	odd	yes	α	1.6E+5 years	6.6	2.8	
^{234}U	^{240}Pu	^{246}Cm	+2p,+0n	even	even	not	α	2.5E+5 years	37	1.4	
^{235}U	^{241}Pu	^{247}Cm	+2p,+1n	even	odd	yes	α, β^-, α	7E+8 years	1E-6	45 (1:3)	c
^{236}U	^{242}Pu	^{248}Cm	+2p,+2n	even	even	not	α	2.3E+7 years	62	1.1	d
^{237}U	^{243}Pu	^{249}Cm	+2p,+3n	even	odd	–	β^-	7 days	32	4.6	
^{237}Np	^{243}Am	^{249}Bk	+3p,+2n	odd	even	not	α, α, β^-,	2.1E+6 years	291	8152	e
^{238}Np	^{244}Am	^{250}Bk	+3p,+3n	odd	odd	–	β^-	2 days	4.8	3.1	
^{238}Pu	^{244}Cm	^{250}Cf	+4p,+2n	even	even	not	α	88 years	4.8	0.02	f
^{239}Pu	^{245}Cm	–	+4p,+3n	–	–	–	–	–	–	–	g

[a]Simplified statement about fission probability by thermal neutrons.

[b]In these cases, the pairing term shifts the balance between Coulomb and asymmetry terms to the right from the stability line. ^{232}Th, ^{238}U and ^{244}Pu belong to the top four actinides with longest half-lives (see Fig. 1).

[c]These nuclides are close to the stability line. However, their behavior is irregular. ^{235}U and ^{247}Cm, as the third and sixth most stable actinide nuclides decay through α decay. Nevertheless, ^{241}Pu decays through β^- and its half-life is by 6 orders of magnitude lower than for ^{235}U. This nuclide causes the major irregularity between the ^{232}Th and ^{238}U irradiation chains.

[d]^{248}Cm can already undergo spontaneous fission.

[e]^{249}Bk decays already by β^-.

[f]^{250}Cf is more stable than ^{244}Cm.

[g]Because of the repetitiveness, refer to the previous occurrence.

Neutron capture rate

The most trivial estimate of the neutron economy is represented by the simple sum of all capture rates in the irradiation chain. This sum can be compared with the average number of neutrons per fission $\bar{\nu}$, reduced by one neutron needed for next fission, to obtain the respective neutron excess or deficiency. It is only an approximate value, its accuracy can be increased by accounting also for the (n,2n) transmutation reaction, which is a source of extra neutrons.

Costs of synthetic actinides fission

Alternatively, the neutron economy can be assessed from the knowledge of the fission rate distribution between the nuclides in the irradiation chain. Each synthetic actinide in the irradiation chain is originated by consecutive transmutations of the primordial nuclide ^{232}Th or ^{238}U. Accordingly, the neutron costs of its creation can be obtained as the difference between its nucleon number and the original nucleon number (232 or 238). The ^{241}Pu creation in ^{238}U irradiation chain costs 3 neutrons (241–238 = 3). Therefore, 4 neutrons are at the end needed for its fission (see **Table 2**). For ^{237}Np in the same chain −1 neutron is needed for creation (237–238 = −1). The cost for its fission is thus 0 neutrons. Based on the knowledge of the fission rate distribution within the irradiation chain the total neutron costs can be obtained. This method is also only approximate. Since many of the nuclides are decaying by α decay, the four nucleons lost in each decay should be accounted for in such a balance.

Neutron balance of the irradiation chain

The overall neutron balance n_{B1} of the actinides chain can be obtained from the average number of neutrons per fission $\bar{\nu}$, reduced by one neutron needed for next fission, and the sum of normalized capture rates C_i and $(n,2n)_i$ rates of a given nuclide i:

$$n_{B1} = \bar{\nu} - 1 - \sum_i C_i + \sum_i (n,2n)_i$$

An alternative neutron balance n_{B2} can be estimated from the fission rate distribution F_i between the nuclides in the irradiation chain and from the normalized α decay reaction rate:

$$n_{B2} = \bar{\nu} - \sum_i F_i n_i - 4\sum_i \alpha_i$$

Table 2 Neutron cost of creation and fission for selected actinide nuclides in ^{232}Th and ^{238}U irradiation chains.

Nuclides chain							
1	*2*	*Position*	*Protons (p)*	*Neutrons (n)*	*Fissile by thermal neutrons*	*Absolute neutron cost to create*	*Absolute neutron cost to fission*
^{232}Th	^{238}U	+0p,+0n	even	even	not	0	1
^{233}U	^{239}Pu	+2p,-1n	even	odd	yes	1	2
^{234}U	^{240}Pu	+2p,+0n	even	even	not	2	3
^{235}U	^{241}Pu	+2p,+1n	even	odd	yes	3	4
^{236}U	^{242}Pu	+2p,+2n	even	even	not	4	5
^{237}Np	^{243}Am	+3p,+2n	odd	even	not	5	6
^{238}Pu	^{244}Cm	+4p,+2n	even	even	not	6	7
^{239}Pu	^{245}Cm	+4p,+3n	even	odd	yes	7	8
^{240}Pu	^{248}Cm	+4p,+4n	even	even	not	8	9
^{241}Pu	^{247}Cm	+4p,+5n	even	odd	yes	9	10
^{245}Cm	^{251}Cf	+6p,+7n	even	odd	yes	13	14

where n_i is the number of neutrons needed for fission of nuclide i (see Table 2) and the product $F_i n_i$ represents the fission costs, neutron costs to create and fission the nuclide. The trivial neutron balance for the four irradiation chains in Figs. 3–6 is illustrated in Table 3. Here it must be stressed, that the result in Table 3 refer only to the neutron balance of the actinides. The overall neutron economy of a reactor will depend also on the capture rate of structural materials, coolants and moderators and on the neuron leakage from the reactor core. Nonetheless, some basic statements can be made, based on the results in Table 3.

The ^{232}Th irradiation chain provides less neutrons per fission, around 2.5 neutrons. At the same time, its neutron capture rate is smaller. Hence it provides neutron excess in both thermal (0.12 neutrons) and fast (0.37 neutrons) spectra. The ^{238}U irradiation chain provides more neutrons per fission, around 2.9 neutrons. However, also the neutron capture rate is higher. The overall neutron excess is positive only in the fast spectrum (0.68 neutrons). In the thermal spectrum there is neutron deficiency (−0.18 neutrons) and the reactor cannot be operated with this fuel composition. Hence, self-sustaining breeding is not possible in thermal spectrum with ^{238}U. The major weakness of the ^{238}U irradiation chain is the lower fission rate of ^{239}Pu. Its major advantage, the higher average number of neutrons per fission, cannot compensate it in thermal spectrum. In ^{238}U uranium chain in graphite moderated MSR 1.08 and 0.54 neutrons are lost by ^{241}Pu and ^{245}Cm fission respectively. The capture rate of ^{239}Pu (35.8%), as a gateway for higher isotopes build-up, is 3× higher than the ^{233}U capture rate (10.6%) in the same reactor.

Radiotoxicity of the irradiation chain

Actinide alpha decay series

Synthetic actinide nuclides, in the discharged fuel from an open cycle practiced today or as the recycling losses in a closed cycle cause a long-term stewardship burden. They have very long half-lives (see Fig. 1) and the daughter products are also radioactive. All nuclides relevant for nuclear fuel cycle undergo either α or β^- decay. Alpha decay reduces number of nucleons by 4 whereas beta decay does not change it. This is the reason why all actinides decay in four separate decay series, which differ by number of nucleons i ($4i$, $4i+1$, $4i+2$, and $4i+3$). These actinide alpha decay series are called: Thorium (4i), Neptunium (4i + 1), Uranium (4i + 2) and Actinium (4i + 3) series and they end with the first available stable nuclide, i.e., ^{208}Pb, ^{209}Bi, ^{206}Pb, and ^{207}Pb. The radiotoxicity should account for the contribution of all the nuclides in the entire decay series.

Since actinides with odd proton number are very unstable, especially when also having odd neutron number; practically all fissile nuclides are in Neptunium and Actinium series. The ^{232}Th and ^{238}U irradiation chains are shifted by 6 nucleons (+2p, +4n); hence, the nuclides in the same position within each chain decay through different series. The pairing term of the Weizsäcker formula cause some irregularity; however the half-lives for radioactive decay generally decrease with increasing nucleon number. The nuclides in the ^{238}U irradiation chains have thus shorter half-lives (see Table 1). At the same time, nuclides from this chain often decay through one of the ^{232}Th irradiation chain nuclides. Finally, evaluation of radiotoxicity is complex, because slowly decaying nuclide, e.g., ^{233}U, can provide low initial radiotoxicity; however, secondary peak can occur once the concentration of daughter product, e.g., ^{229}Th, will be stabilized. To illustrate this behavior, the radiotoxicity of selected nuclides caused by 10^{30} atoms is shown in Fig. 8. It represent the radiotoxicity caused by ingestion of the radioactive materials by human. The necessary coefficients have been adopted from (Soti et al., 2019). This figure shows radiotoxicity normalized to 10^{30} atoms. Accordingly, the values for some minor nuclides are exaggerated. However, it is identifying nuclides with substantial secondary peak: ^{233}U (^{233}Th), ^{234}U (^{238}Pu, ^{242}Am, ^{242m}Am), and ^{237}Np (^{241}Pu). The case of ^{237}Np is particular because it decays through ^{233}U; however, it has long half-live, which delays the ^{233}U secondary peak occurrence. It is obvious that nuclides with secondary peak are predominantly located in the ^{232}Th irradiation

Table 3 Neutron cost of creation and fission of selected actinide nuclides in ^{232}Th and ^{238}U irradiation chains.

	^{232}Th chain	*^{233}U*	*^{235}U*	*^{239}Pu*	*^{241}Pu*	*^{245}Cm*	*Sum fissile*	*^{232}Th*	*^{234}U*	*^{236}U*	*^{237}Np*	*^{238}Pu*	*Sum fertile*	*Sum others*	*Sum total*	$\bar{\nu}$	n_{B1} -n_{B2}
	Fissile	Y	Y	Y	Y	Y	–	N	N	N	N	N	–	–	–		
Graphite moderated MSR	Capture rate %	10.6	2.46	0.83	0.24	0.03	14.2	99.1	13.3	2.40	2.35	2.14	119	4.81	138	2.50	0.12
	Fission rate %	85.6	10.9	1.36	0.61	0.18	98.7	0.76	0.19	0.06	0.01	0.12	1.15	0.17	100		–
	Fission cost	1.71	0.44	0.11	0.06	0.02	2.34	0.01	0.01	0.00	0.00	0.01	0.03	0.01	2.38		0.12
MCFR	Capture rate %	9.53	1.10	0.00	0.00	0.00	10.6	96.9	5.47	0.85	0.66	0.21	104	1.45	116	2.53	0.37
	Fission rate %	86.4	4.37	0.18	0.01	0.00	90.9	2.76	5.15	0.24	0.19	0.42	8.76	0.30	100		–
	Fission cost	1.73	0.17	0.01	0.00	0.00	1.92	0.03	0.15	0.01	0.01	0.03	0.23	0.00	2.16		0.37
	^{238}U chain	^{239}Pu	^{241}Pu	^{245}Cm	^{247}Cm	^{251}Cf	Sum fissile	^{238}U	^{240}Pu	^{242}Pu	^{243}Am	^{244}Cm	Sum fertile	Sum others	Sum total	$\bar{\nu}$	n_{B1} - n_{B2}
	Fissile	Y	Y	Y	Y	Y	–	N	N	N	N	N	–	–	–		
Graphite moderated MSR	Capture rate %	35.8	10.2	1.01	0.30	0.11	47.5	95.5	37.8	10.2	10.1	7.80	161	3.46	212	2.95	−0.18
	Fission rate %	60.0	27.1	6.79	0.61	0.23	94.7	4.28	0.14	0.15	0.05	0.40	5.03	0.31	100		–
	Fission cost	1.20	1.08	0.54	0.06	0.03	2.92	0.04	0.00	0.01	0.00	0.03	0.09	0.02	3.02		−0.17
MCFR	Capture rate %	18.3	1.34	0.07	0.01	0.00	19.8	88.8	11.0	1.02	0.94	0.42	102	2.85	125	2.93	0.68
	Fission rate %	70.8	7.74	0.35	0.04	0.00	78.9	10.91	7.75	0.56	0.11	0.23	19.6	1.50	100		–
	Fission cost	1.42	0.31	0.03	0.00	0.00	1.76	0.11	0.23	0.03	0.01	0.02	0.39	0.03	2.18		0.68

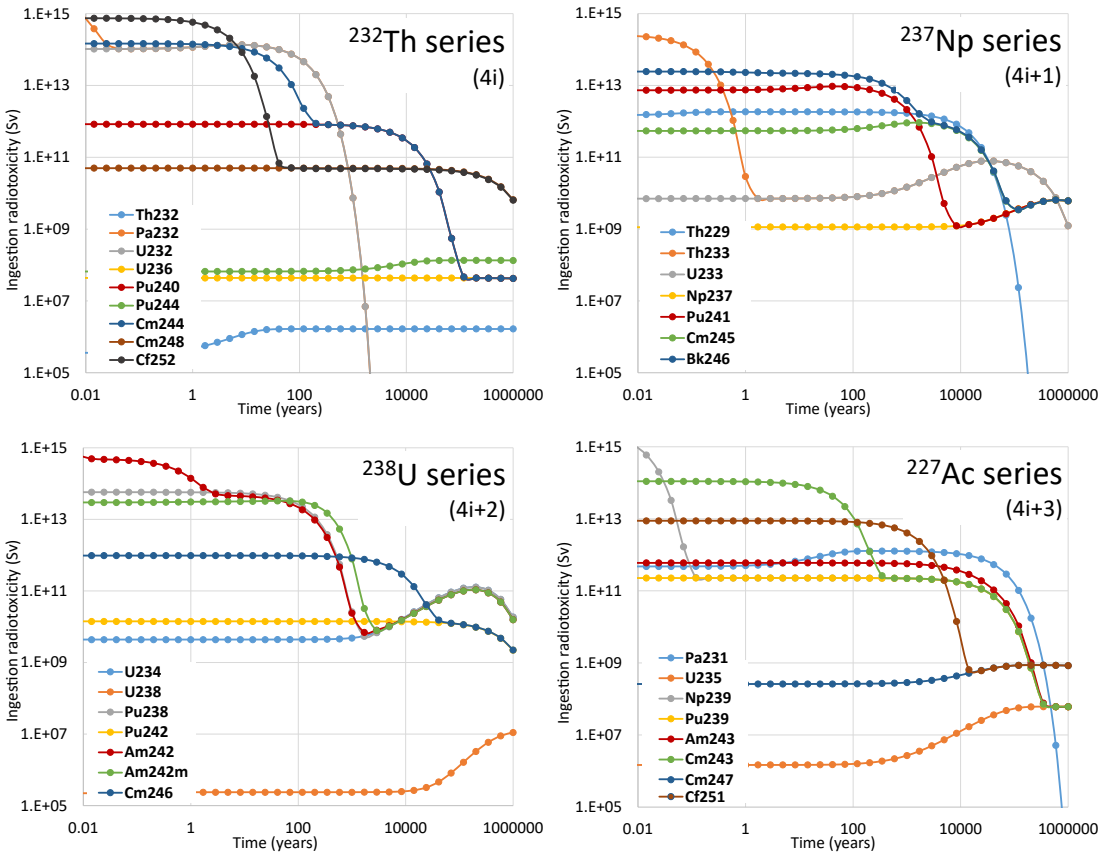

Fig. 8 Evolution of ingestion radiotoxicity originated from 10^{30} atoms of selected nuclides in the Thorium (top-left), Neptunium (top-right), Uranium (bottom-left) and Actinium (bottom-right) series.

chain. For ^{235}U, ^{238}U, and ^{244}Pu, there is a late radiotoxicity increase caused by longer half-live of the daughter products ^{231}Pa, ^{234}U, and ^{240}Pu. However it is rather plateau than secondary peak. Similar radiotoxicity increase appears earlier for ^{232}Th, because its daughter product ^{238}Th has relatively short half-live. In all these cases, the radiotoxicity level is low and, with exception of ^{244}Pu related to the natural ore radiotoxicity.

Radiotoxicity of the ^{232}Th and ^{238}U irradiation chains

The ingestion radiotoxicity of 10^{30} atoms can be presented also for the actinides compositions from the four irradiation chains presented in Figs. 3–6. In the ^{232}Th irradiation chain, actinides composition in thermal spectrum provide slightly lower radiotoxicity. This is caused by lower absolute share of synthetic actinides. The two curves follow the same trend. In both cases a secondary peak occurs (see left Fig. 9). The radiotoxicity of ^{238}U irradiation chain is initially higher in thermal spectrum. The absolute share of synthetic actinides is lower in thermal spectrum also in this case. However, the difference is smaller and the thermal spectrum chain includes larger share of higher synthetic actinides nuclides. Since these decay faster, already earlier than 100 years after discharge the fast spectrum chain provides higher radiotoxicity. The secondary peak is absent in ^{238}U chain and the radiotoxicity continuously decreases (see right Fig. 9).

The results from Fig. 9 can be used to compare the relative radiotoxicity of the ^{232}Th and ^{238}U irradiation chains. Their ratio is presented in left Fig. 10 using logarithmic time scale and in right Fig. 10 using linear time scale. As expected, ^{232}Th chain provides initially up to 40 × lower radiotoxicity. Nonetheless, at longer time periods the secondary peak results in up to 20 × higher radiotoxicity. Accordingly, the final conclusion would be determined by time period of interest.

Synthetic actinides from the irradiated fuel become waste once they cannot be recycled and reused. In the open cycle they become waste by political decision not to recycle and reuse them. In the closed fuel cycle it is rather the recycling frequency and technology efficiency. There will be always some fraction of synthetic actinides in reprocessing losses. Therefore, the maximal resources utilization depends on the reprocessing losses as shown by Fig. 2. The radiotoxicity of synthetic actinides per unit of produced energy in the closed cycle depends on the resources utilization level. To provide an example, the initial radiotoxicity, just after shut down of the equilibrium reactor, per unit of produced energy is plotted in left Fig. 11 as a function of natural resources utilization. The radiotoxicity per unit of energy linearly decrease with increasing resources utilization. However, the

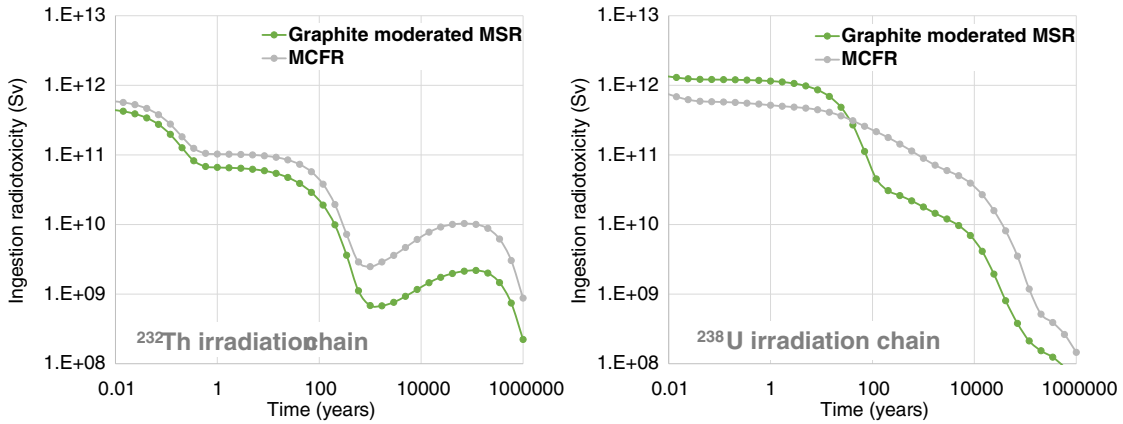

Fig. 9 Evolution of ingestion radiotoxicity originated from 10^{30} atoms of the ^{232}Th (left) and ^{238}U (right) irradiation chains in the graphite moderated MSR and MCFR.

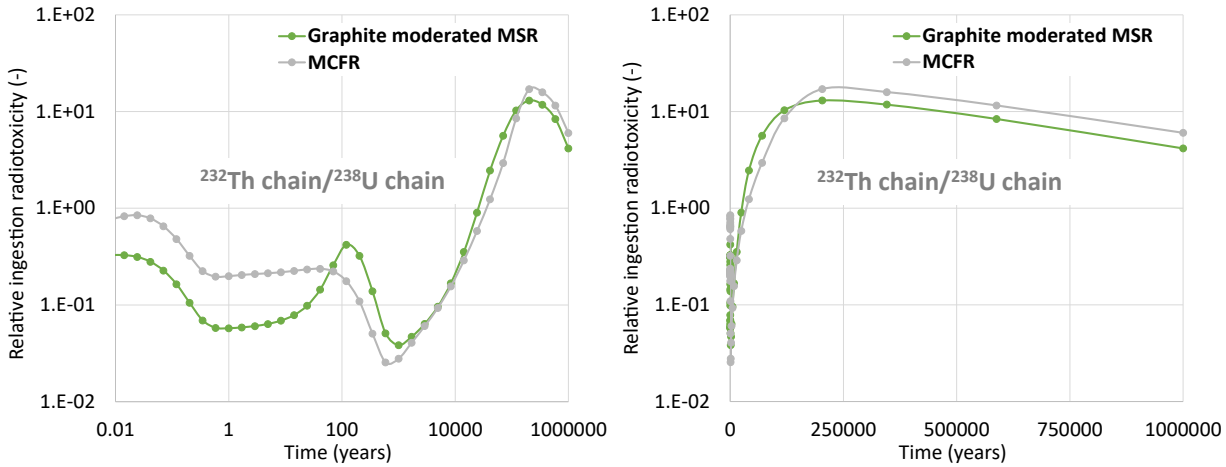

Fig. 10 Evolution of the ratio between the ingestion radiotoxicity in ^{232}Th and ^{238}U irradiation chains in logarithmic (left) and linear (right) time scale.

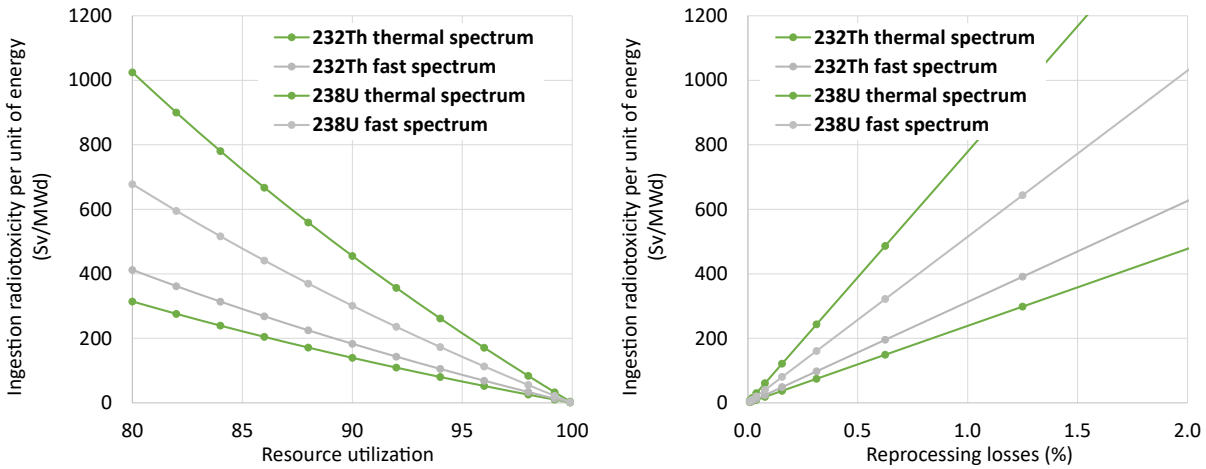

Fig. 11 Dependency of the ingestion radiotoxicity per unit of produced energy on resources utilization (left) and reprocessing losses (right) for ^{232}Th and ^{238}U irradiation chains.

utilization depends on reprocessing losses and reprocessed fuel burnup in FIMA %. Hence, in the right Fig. 11 the same initial radiotoxicity per unit of energy is plotted as a function of reprocessing losses, assuming 5% FIMA burnup of the reprocessed fuel. Since the burnup is fixed in this example, the radiotoxicity per unit of produced energy grows linearly with increased reprocessing losses.

Summary

The nuclear fuel cycle relies on resources, which are not renewable. To fully utilize their potential self-sustaining breeding in closed cycle is needed, which is demanding from the neutron balance perspective. Not all reactor concepts are capable to achieve self-sustaining breeding and fully utilize the natural resources. Yet, many advanced reactors are capable of self-sustaining breeding and thus to produce energy from primordial ^{232}Th and ^{238}U nuclides, using synthetic fissile nuclides ^{233}U and ^{239}Pu as a kind of catalyzer. The fuel composition in a self-sustaining breeder operated in a closed cycle tends to converge to equilibrium composition. The type of irradiation chains and the reactor neutron spectrum determine the equilibrium composition.

These chains follow the line of stability and show a certain symmetry. This symmetry is explained in this article by the binding energy per nucleon in each of the chain nuclides. The properties of actinides nuclides are repeating in the nuclide chart with the step of 2 protons and 4 neutrons. The major difference between these chains is that almost all nuclides in the ^{232}Th irradiation chain have longer half-life for radioactive decay than their respective nuclides in the ^{238}U chain. This is an advantage from several perspectives. The available ^{232}Th resources are larger than those of ^{238}U thanks to the longer half-life of ^{232}Th. By the same token, the stewardship burden caused by high-level waste of the ^{232}Th irradiation chain is smaller in the short term. However, it also results in 11 times higher ^{233}Pa concentration than the respective ^{239}Np concentration. The increased parasitic neutron capture probability of ^{233}Pa slightly deteriorates the neutronic performance of ^{232}Th irradiation chain. Another difference is the irregular behavior of ^{241}Pu in the ^{238}U chain relative to ^{235}U, which is at the respective position in ^{232}Th irradiation chain: ^{241}Pu has a six orders of magnitude shorter half-life and decays by β^- decay, versus alpha decay of ^{235}U. The last important difference is that ^{233}U and ^{235}U as the major fissile nuclides in ^{232}Th chain have lower number of nucleons than the respective ^{239}Pu and ^{241}Pu in the ^{238}U chain, and therefore release, on average, less neutrons per fission.

The ^{232}Th and ^{238}U irradiation chains are presented in thermal and fast reactor spectra. Based on the nuclide concentrations and reaction rates, neutron economy is quantified and long term ingestion radiotoxicity assessed. The existing knowledge was confirmed that self-sustaining breeding in closed cycle is not possible in thermal spectrum reactors when fed with ^{238}U and questionable even when fed with ^{232}Th. In fast neutron spectrum the ^{238}U irradiation chain profits more from neutron spectrum hardening and performs better than the ^{232}Th irradiation chain.

The ingestion radiotoxicity is lower for ^{232}Th irradiation chain on relatively short time scale but higher on the long time scale. The longer half-lives provide initial advantage to the ^{232}Th irradiation chain, but this chain features a radiotoxicity peak several 10s of 1000s years following discharge. Beyond $\sim$20,000 years the ^{238}U irradiation chain radiotoxicity is lower due to the shorter half-lives of its nuclides.

The general conclusion of this article is that self-sustaining breeding in closed cycle can be achieved with both primordial nuclides ^{232}Th and ^{238}U. The differences between the respective irradiation chains are relatively small and both nuclides represent valuable natural resource.

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